Reply to reviewer 1

Meteorological factors controlling low-level continental pollutant outflow across a coast

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We would like to thank the author for his/her comments on the paper.

Main Points

1. page 10854, line 26: I dont understand why this definition of coastal outflow does not include transport within the marine boundary layer. I realize that pollution in warm continental air will rise above cold air in the MBL when blown out to sea. But if a cold front advects from land out to sea the pollution is emitted into very cold air over land. When this cold polluted air mass moves over the relatively warmer waters of the Gulf Stream along the US east coast an unstable situation arises. As a result, mixing occurs from the surface of the ocean to the top of the convective clouds that are typically produced in this scenario (this truly is a common occurrence along the US east coast in winter and spring). Furthermore, Lee et al. (2011) show CO in excess of 200 ppbv at altitudes less than 500 m downwind of New York City, which is pollution that advected offshore within the MBL.

Lee, S.-H., et al. (2011), Modeling ozone plumes observed downwind of New York City over the Nor th Atlantic Ocean during the ICARTT field campaign, Atmos. Chem. Phys., 11, 73757397.

In the paper, the term coastal outflow is used to describe the decoupling of pollution from the surface via the formation of an internal stable boundary layer which occurs when there is horizontal transport from land to sea and the land boundary layer is deeper than the marine boundary layer. We focus on summer days when the SST off East Coast USA is much lower than land surface temperature and the marine boundary layer is stable (especially across the Gulf of Maine which was the focus of the ICARTT observational experiment). We have included this more precise definition of coast outflow in the paper to emphasize the importance of decoupling in this process and it's distinction from other ventilation mechanisms such as occurring during the passage of fronts.

Also, the new Fig.5d showing aircraft profiles of ozone illustrates the difference in chemical evolution for air travelling in the marine boundary layer and the air above it. Above the boundary layer the pollution signature can be transported for much longer times and greater distances.

We have also used the analysis of Lee et al (2011) in much more detail in Section 2.2 of the revised paper.

2. A paper that needs to be referenced is Fang et al. (2009) which examines 15 summers of pollutant outflow from the eastern USA:

Fang, Y., et al. (2009), Estimating the contribution of strong daily expor

t events to total pollutant export from the United States in summer, J. Geophys. Res., 114, D23302.

We have included reference to Fang et al. 2009 in the introduction. Thankyou for pointing it out.

3. Figure 4 compares the modeled surface tracer (emitted uniformly from the land surface) to CO measurements made at 121 EPA stations across the eastern USA. I find this comparison to be essentially meaningless and suggest a more appropriate measurement/model comparison method. The modeled tracer is released uniformly across the eastern North America land surface which is predominantly rural. In contrast the C2687 CO monitors are mainly in urban areas, which is evident from the very high mean CO mixing ratios of 400-600 ppby. These EPA monitors are only useful in urban areas because their poor detection limit is about 200 ppby, higher than typical rural CO mixing ratios. I realize that the authors are not trying to exactly reproduce CO mixing ratios, but the very high modeled CO mixing ratios demonstrate that the emission rate needs to be reduced. Seeing as the goal of the study is to understand export out of the boundary layer a much better comparison is to compare the vertical distribution of the modeled tracer to vertical profiles of measured CO. For example: 1) Fang et al. [2009] show in their Figure 2 mean CO mixing ratio profiles as measured by the NASA DC8 during ICARTT. 2) Lee et al. [2011] compare WRF-Chem to NOAA P3 CO profiles downwind of New York City on several days during ICARTT. 3) Cooper et al. [2006] show CO profiles above Texas, Atlanta, the northeastern USA and Montreal Canada during ICARTT. These data are freely available from the IAGOS/MOAZIC database: www.iagos.org The authors need to obtain the CO data from the NASA DC8, NOAA P3 and MOZAIC/IAGOS and produce a composite CO profile above the eastern USA and above the western North Atlantic Ocean. They then need to see if their modeled tracer has the same general shape as the measured profile, examining the model standard deviation as well as the mean. They also need to carefully compare their tracers vertical distribution to the CO profiles from the NOAA P3 flights downwind of New York City on July 20-21, 2004 as described by Lee et al. [2011]. This is the only way to see if the model provides a realistic simulation of offshore transport.

Cooper, O. R., et al. (2006), Large upper tropospheric ozone enhancements above midlatitude Nor th America during summer: In situ evidence from the IONS and MOZAIC ozone measurement network, J. Geophys. Res., 111, D24S05, doi:10.1029/2006JD007306.

Thanks for the suggestion to make more of the aircraft data and in particular the ITCT-Lagrangian case and the link with Lee et al (2011). We have re-written Section 2.2 which compares the idealised tracer simulations with observations from ICARTT. The new Figure 5a shows vertical profiles of tracer mixing ratio from UM simulation on the 20th, 21st and 22nd July. Profiles are averaged over sea points only and in areas corresponding to those flight segments of the NOAA WP-3D aircraft tracks that are linked in a Lagrangian fashion (shown on Fig. 1). This enables us to compare and contrast the shape of the vertical profiles and their evolution following an air mass over the sea moving away from coastal emissions. All of the profiles demonstrate a similar shape with the highest tracer mixing ratios found below 700m in the top of the marine boundary layer. Above 1km the tracer mixing

profiles decrease roughly exponentially to 4km. Although retaining similar profile shape, the magnitude of the mixing ratios reduces with time.

Figure ??(b)-(d) show the ICARTT aircraft observations of CO, NOx and ozone respectively. The shape and evolution of tracer is consistent with the observed CO, given that CO has a much longer photochemical lifetime than the decay timescale of the tracer. NOx shows similar behaviour but the profile slope is influenced by photochemistry. As expected, ozone does not have a profile consistent with a primary pollutant. It serves to illustrate the difference between the evolution of outflow in the MBL and the layer above.

We believe that the regional comparison of surface CO with the domain integral tracer over the continental boundary layer is meaningful, since the domain integral of tracer must reflect the sum of sources emitted into it (plus advection from the far field). Although the EPA sites are in urban areas with stronger emissions, this is fine since they must therefore dominate the actual CO burden over the East Coast. However, we have omitted the figure since the use of the aircraft data serves more effectively to demonstrate the performance of the tracer simulation.

In contrast with the surface sites and their continuous time series, we did not think that composites of aircraft profiles from across the eastern USA would be a particularly good comparison to make. The range of mixing ratios in the air masses encountered on profiles of the NASA DC8 and NOAA WP-3D is massive, in part reflecting the spatial and temporal variability of emissions, as well as long-range transport (for example the boreal forest fire emissions which were a feature of the ICARTT period). Even with the MOSAIC flights the dataset is sparse in time. Instead we chose to analyse the ITCT-Lagrangian case in more detail since there is a clear rationale for linking profiles, and those profiles are over the sea away from localised emissions.

4. The authors need to add a figure that shows the percent of tracer advected horizontally into the ocean each day, along with the percent transported vertically. They also need to add discussion of these budget terms that are presently only briev mentioned in the conclusions.

We have included the new Figure 7 which shows the 24 h running average timeseries of the mass transport of tracer as a percentage of total mass of tracer. The figure shows that whilst the flux from land to the marine boundary layer is small the flux from land to the coastal outflow layer is large and comparable to the flux from land to the free-troposphere layer over land.

Minor Points

- 1. page 10854, line 6 and quantifies their importance Corrected.
- 2. page 10854, line 18 The abstract would flow better if the sentence beginning with Short-lived tracers came before the sentence on line 16 that begins with For short-lived tracers Corrected.
- 3. **p10857 line 15 that have the most influence** Corrected.
- 4. **p10857 line 19 North Atlantic Ocean** Corrected.

- 5. **p10858 line 14 cumulus-capped** Corrected.
- 6. p10858 line 15 I dont understand why the number to turbulent mixing levels would be capped at the cloud base. A convective cloud has vertical mixing from the cloud base to the top of the cloud so I dont see how you can cap NTML at cloud base.

The NTML is not "capped at cloud base". The Richardson-number based turbulent boundary layer scheme tends to identify the top of the boundary layer with the top of the "mixed layer" in a neutral or convective situation. In the case of shallow moist convection, it turns out that this level is at the base of the stratocumulus deck and the cloud layer itself is partially mixed. This also happens in the ECMWF model. In the MetUM, above the NTML the convection scheme mixes tracer from cloud base to the top of the cloud (below this level mixing is performed by the boundarylayer scheme). So, although it is partitioned between schemes in the model, tracer is mixed across this region. We have added a sentence to the paper to clarify this.

7. p10858 line 20 Hmax has a single value determined from all the daily values, so how can Hmax vary from day to day when it has a single value?

We have re-written this sentence to clarify how H_{max} was calculated. The text now reads 'The maximum boundary layer height at each land point every day was calculated. The 90th percentile of the maximum boundary layer height was found to vary little from day to day, thus H_{max} was fixed at 2000m. The residual layer is defined to extend from the top of the boundary layer to H_{max} .

8. p10858 line 21 The free troposphere must, by definition, only extend to the tropopause but you state that it extends to the top of the model which is located at 39 km (near the top of the stratosphere). The calculations have been performed from the top of the residual layer to the top of the model (39km). However, in practice, almost no tracer penetrates into the stratosphere and so the results are unlikely to change if we performed the calculation from the top of the residual layer to the tropopause. We agree that our use of the term 'free troposphere' is therefore not strictly correct, but in this case we feel that it adequately reflects what happens in practice. The text has been editted to make this clear.

- 9. page 10860 line 11 importance of convection can be quantified. Corrected.
- 10. page 10863 line 11 minus the tracer that excluded the convective mass Corrected.
- 11. page 10865 line 20 maximum height of the continental Corrected.
- 12. page 10868 line 6 an analytic solution Corrected.